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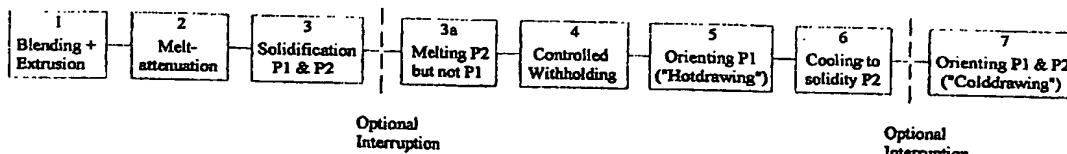
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(54) Title: METHOD OF MANUFACTURING ORIENTED FILMS FROM ALLOYED THERMOPLASTIC POLYMERS, APPARATUS FOR SUCH MANUFACTURE AND RESULTING PRODUCTS



(57) Abstract: A blend of two partly incompatible polymers is extruded to form a film under conditions such that the alloy in the blend comprises fibrils extending mainly in one direction, the width and thickness of each fibril having a mean lower than 5 μm , the fibrils being formed of the polymer component having a higher melting point, and the fibrils being surrounded by a matrix of the second polymer, which has a lower coefficient of elasticity. The special morphology is achieved by providing grids (301, 302) in a grid chamber upstream of the exit orifice of the extrusion die, and by sequential hot stretching at a temperature at which the higher melting polymer is at least partly crystalline, and cold stretching, at which both polymers are substantially solid. The second, cold stretching step, is carried out under conditions to give an elongation at break at 20°C of at least 25%. The films are of particular value for forming crosslaminate having high shock resistance. Preferably the polymer forming fibrils is polypropylene and the polymer forming the matrix is an ethylene polymer, preferably LLDPE.

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AMENDED CLAIMS

[received by the International Bureau on 10 November 2004 (10.11.04);
original claims 1-88 replaced by amended claims 1-97 (15 pages)]

1. A method of manufacturing an oriented film by forming a blend of at least two polymers P1 and P2, which both are at least partly crystalline at a temperature below 100°C, whereby P1 has a mechanically determined melting point which is at least 20°C higher than the mechanically determined melting point of P2, extending the blend to form a film and stretching the film, whereby the polymers are selected such that they exist as separate phases in the stretched film, and P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1, and
5 oriented film comprises an alloy of the polymer which is a dispersion of fibrils of P1 surrounded by P2, whereby each fibril extend mainly in one direction and generally has width and thickness which as a mean of these two dimensions is around or lower than 5µm, and further stretching of the film partly takes place by draw-down after extrusion of film while both
10 components are at least partially molten, and partly at a later stage
15 characterised in that the film after said draw-down is hot stretched while P1 is in solid state and P2 is molten or semimolten to selectively orient P1, such that the elongation at break in the direction of this hot stretching, determined by slow drawing at 20°C, is at least 25%, this hot stretching being carried out
20 by drawing the film over a frictionally withholding device.
2. A method according to claim 1, characterised in that after said hot stretching the film is further stretched while both components P1 and P2 are solid, preferably in such a manner that the product film has an elongation at break at 20°C (by slow drawing) of at least 25% in any
25 direction.
3. A method according to claim 1 or claim 2, characterised in that the fibrils are flat with average thickness generally around or lower than 1 µm, preferably generally around or below 0.5 µm, and more preferably generally around or lower than 0.1 µm, and with average width generally
30 around or lower than 5 µm.

4. A method according to claim 1, characterised in that in order to reduce the cross dimensions of the fibrils, the molten blend during extrusion is passed through at least one screen or grid located in a chamber immediately upstream of the exit orifice of the extrusion device, said 5 chamber having a gap higher than the gap of the exit orifice.

5. A method according to claim 4, characterised in that each such grid has walls extending several millimetres in the direction of the flow of the molten blend.

6. A method according to claim 5, characterised in that the major 10 walls in each such grid are slanted so that each forms an angle between about 10 to about 70° to the major surface of the flow entering the grid.

7. A method according to claim 6, characterised in that the slanting and the wall thickness and distances between the walls are such that in a longitudinal section of the device perpendicular to major surface of 15 the flow as this enters the grid, there are at least four such walls.

8. A method according to claim 6, characterised by at least two grids, where in the such walls of one grid are slanted in the opposite direction to the walls of the other grid.

9. A method according to claim 1, characterised in that in 20 succession to the extrusion and attenuation of the blend while both P1 and P2 are molten, the film is first cooled to solidify P1 and P2, thereafter the film is heated in air-lubricated engagement with a heating body of controlled temperature to melt or at least partially melt P2, while keeping P1 solid, and immediate thereafter, while P2 still is at least partially molten and P1 is solid, 25 the film is subjected to the said selective orientation of P1 and subsequent solidification of P2.

10. A method according to claim 1, characterised in that the frictional withholding device comprises one or more bars with rounded edges over which the film is dragged while following an adjustable arc of the bar 30 edge, and said bar or bars are maintained at a temperature which prevents

the film from sticking to the edge or edges, and the length of travel in contact with the edge or edges is adapted to prevent P2 from wholly solidifying.

11. A method according to claim 9, characterised in that at least the process steps from and including extrusion to and including the solidification of P2 are carried out in-line, whereby the line also comprises a hold-back device acting between the cooling and the subsequent heating, and preferably the process steps following solidification of P2 are also carried out in-line with the former process steps.
12. A method according to claim 11, characterised in that the film is extruded as a flat film, and the controlled hold-back between cooling and subsequent heating is established by a roller arrangement, which also may supply the said cooling.
13. A method according to claim 11, characterised in that the film is formed and treated in tubular form from extrusion and at least to the final solidification of P2, whereby the controlled hold-back between cooling and subsequent heating is established by one or more circular rings with rounded edges over which the film is dragged while following an adjustable arc of the rounded edge, and said ring or rings are maintained at a temperature which prevents the film from sticking to the said edge or edges.
14. A method according to claim 11, characterised in that the said heating is carried out with the film in air-lubricated engagement with two heating bodies of which is provided one on each side of the film, the spacing between said heating bodies preferably being adjustable.
15. A method according to claim 1, characterised in that the film immediately after the extrusion is cooled to solidification of P1 while P2 is kept molten or semimolten, and further in immediate succession, the selective orientation of P1 over a frictionally withholding device is carried out with the polymers in such states.
16. A method according to claim 15, characterised in that the frictionally withholding device comprises one or more bars with rounded edges over which the film is dragged while following an adjustable bow-

length of the edge, and said bar or bars and the length of travel in contact with the edge or edges is adapted to prevent P2 wholly solidifying.

5 17. A method according to claim 15, characterised in that the cooling to the said state is carried out by air-lubricated engagement of the film with a cooling body of controlled temperature.

18. A method according to claim 17, characterised in that the cooling is carried out with the film in air-lubricated engagement with two heating bodies, one on each side of the film, the spacing between said heating bodies preferably being adjustable.

10 19. A method according to claim 2, characterised in that the said further stretching is carried out in the same longitudinal direction as the hot stretching of the film.

15 20. A method according to claim 19, characterised in that by a suitable selection of the conditions for the different stretching processes, and optionally by addition of a finely dispersed fracture-promoting material to the extruded blend, the longitudinal orientation after full solidification is adapted to produce locations of rupture of the P1 fibrils and in connection with such rupture extra orientation of P2 in and around the said locations, the locations being generally extended in a linear fashion at an angle to the 20 direction of orientation.

21. A method according to claim 19, characterised in that the said further stretching is carried out at around 50°C or at a lower temperature.

25 22. A method according to claim 19, characterised in that in succession to said further stretching, transverse stretching is carried out while P1 and P2 are solid, preferably under allowance of a simultaneous longitudinal contraction.

30 23. A method according to claim 22, characterised in that longitudinal contraction is achieved by forming transverse pleats in the film prior to the transverse stretching, and the latter is carried out by means of a tenter frame.

24. A method according to claim 2, characterised in that the said further stretching is carried out transversely of the preceding longitudinal orientation of the film, preferably while the film is allowed to shrink in said longitudinal direction.

5 25. A method according to claim 24, characterised in that the shrinking is achieved by forming transverse pleats in the film prior to the transverse stretching, and the latter is carried out by means of a tenter frame.

10 26. A method according to claim 1, characterised in that a minor surface layer is coextruded on at least one side of the blend to enhance bonding properties and/or modify frictional properties of the film.

27. A method according to claim 1, characterised in that P1 consists of polypropylene polyamide or polyethylene terephthalate, and P2 mainly consists of a propylene copolymer or polyethylene.

15 28. A method according to claim 27 in which said polypropylene for P1 is a crystalline copolymer of propylene.

29. A method according to claim 27 or 28 in which the polyethylene is a crystalline copolymer of ethylene, preferably linear low density polyethylene.

20 30. A method according to claim 1, characterised in that after the end of the mentioned steps, the film, which exhibits a uniaxial or unbalanced orientation, is laminated to one or more similarly or differently manufactured films of uniaxial or unbalanced biaxial orientation, whereby the films are arranged so that their main directions of orientation cross each other.

25 31. A method according to claim 1, characterised in that additionally to the mentioned steps the film is cut into narrow longitudinally oriented tapes.

30 32. A method of forming a film or sheet of thermoplastic polymer alloy in which there is formed an intimate blend of polymer material P1' and polymer material P2', the blend is extruded through a die and the extruded film is stretched after extrusion in which the flow passage through the die

comprises an exit orifice having an exit gap, characterised in that upstream from the exit orifice there is provided a grid chamber comprising one or more grids through which the blend passes, the grid or grids having at least 4 (in the longitudinal sections perpendicular to the main surfaces of the flow)

5 closely spaced lamellae having walls extending several millimetres in the direction of the flow, and, between the lamellae apertures of a size selected to reduce the average size of the dispersed phase of P1' or P2' in the blend, the grid or grids being located at a position in the chamber where the gap is wider than the said exit gap, the grid chamber further comprising a gap

10 reduction portion between the screen and the die exit wherein the gap through which the blend flows is reduced at least part way to the gap of the die exit.

33. A method according to claim 32 characterised in that the lamellae in each such grid are slanted so that each forms an angle between about 10 to about 70° to the major surface of the blend flow entering the grid.

15 34. A method according to claim 33, characterised in that the major lamellae in each such grid are substantially planar.

35. A method according to claim 33, in which the lamellae are substantially parallel to the flow as it enters the grid.

20 36. A method according to claim 34, characterised by at least two such grids which mutually are oppositely slanted in relation to the direction of the blend flow entering the grid.

37. A method according to claim 32, characterised in that there is coextruded a surface layer at least on one side of the blend flow, preferably before this flow meets the grid or grids.

25 38. A method according to claim 32 characterised in that P1' and P2' are incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised either by use of an alloying agent or mechanically by sufficient mixing and attenuation, and P2' in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1', and preferably but not necessarily the mechanically

determined melting point of P1' is at least about 20°C higher than that of P2', and further by adaptions of rheological conditions, percentages of the components, and conditions for mixing and extruding a dispersion of microscopically fine fibrils or fibril network of P1' surrounded by P2' is formed

5 in the alloy, whereby each fibril extends mainly in one direction and generally has a thickness around or lower than 5µm, preferably around or lower than 1µm, and still more preferably around or lower than 0.1µm and width at least 5 times its thickness, and further characterised in that the film is stretched after at least P1' has been solidified.

10 39. A method according to claim 38, characterised in that the said stretching is transverse to the direction of the fibrils, and preferably the film is allowed to contract in the direction of the fibrils during said stretching.

15 40. A method according to claim 38, characterised in that possibilities for contraction are introduced by a preceding fine transverse pleating of the film.

41. A method according to claim 40, characterised in that the step of stretching transverse to the direction of the fibrils is preceded by stretching in the direction of the fibrils while the latter are solid.

20 42. A method according to claim 38, characterised in that P1' consists of polypropylene, polyamide or polyethylene terephthalate, and P2' mainly consists of a propylene copolymer or polyethylene.

43. A method according to claim 42 in which the polypropylene for P1 is a crystalline copolymer of propylene.

25 44. A method according to claim 42 or 43 in which the polyethylene is a copolymer of ethylene, preferably linear low density polyethylene.

30 45. A method according to claim 38, characterised in that the film is given a strong uniaxial or unbalanced biaxial orientation, and subsequently the film is laminated to one or more similarly or differently manufactured film of uniaxial or unbalanced biaxial orientation, whereby the films are arranged so that their main directions of orientation cross each other.

46. A method according to claim 38, characterised in that subsequently the film is cut into narrow longitudinally oriented tapes.

47. A method according to claim 32 characterised in that P1' is chosen to exhibit desirable barrier properties, and P1' and P2' are 5 incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised either by use of an alloying agent or mechanically by sufficient mixing and extension, and preferably but not necessarily the mechanically determined melting point of P1' is at least about 20°C higher than that of P2', and further by adaptions of rheological 10 conditions, percentages of the components, and conditions for mixing and attenuation a dispersion of microscopically fine fibrils or fibril network of P1' surrounded by P2' is formed in the alloy whereby each fibril extends in one main direction, has a thickness around or lower than 5µm, preferably around or lower than 1µm, and has a width at least 5 times its thickness.

48. A method according to claim 32 characterised in that P1' and P2' are incompatible to such an extent that they exist as separate phases in the final film, but are compatibilised either by use of an alloying agent or mechanically by sufficient mixing and extrusion and preferably but not necessarily the mechanically determined melting point of P1' is at least 15 about 20°C higher than that of P2', and further by adaptions of rheological conditions, percentages of the components, and conditions for mixing and attenuation a dispersion of microscopically fine fibrils or fibril network of P1' surrounded by P2' is formed in the alloy, whereby each fibril extends mainly 20 in one direction, has a thickness around or lower than 5µm, preferably around or lower than 1 µm, and has width at least 5 times its thickness, and further characterised in that there is added a volatile expansion agent prior 25 to or during the extrusion, which agent is soluble in P2' but generally not in P1', whereby expansion takes place after extrusion.

49. An extruded oriented film which is in the form of a 30 crosslaminate, in which it is laminated to another oriented film, whereby the main directions of orientation cross each other, or is in the form of a rope, twine or woven-tape products, the film comprising a layer of alloy of at least

two polymers P1 and P2, which both are at least partly crystalline at temperatures less than 100°C, wherein P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1, and the alloy comprises a dispersion of microscopically fine fibrils or

5 fibril network of P1 surrounded by P2, wherein each fibril extends mainly in one direction and generally has width and thickness wherein the mean of these two dimensions is around or lower than 5µm, characterised in that

10 a) the P1 fibrils are flat and generally parallel with the main surfaces of the film with thicknesses generally around or lower than 1 µm and width at least 5 times the thickness, and/or

b) the oriented film exhibits locations of rupture of the P1 fibrils, which locations each extend in a generally linear fashion across the film at an angle to the direction of orientation.

15 50. A film according to claim 49, characterised by a minor coextruded surface layer on at least one side of the alloy layer to enhance bonding properties and/or modify frictional properties of the film.

51. A film according to claim 49, characterised in that P1 consists of polypropylene polyamide or polyethylene terephthalate, and P2 mainly consists of a propylene copolymer, or polyethylene.

20 52. A film according to claim 51 in which the polypropylene is a crystalline copolymer of propylene.

53. A film according to claim 51 in which the polyethylene is a copolymer of ethylene, preferably linear low density polyethylene.

25 54. An extruded film according to claim 49 which is a crosslaminate.

55. A film according to claim 49 in the form of rope, twine or woven-tape products.

56. An extruded film comprising a layer of an alloy of at least two polymers P1 and P2, which both are at least partly crystalline at

30 temperatures under 100°C and are incompatible to such an extent that they exist as separate phases in the final film but are compatibilised sufficiently for practical purposes, comprising a dispersion of microscopically fine fibrils

or fibril network of P1 surrounded by P2, wherein each fibril extends mainly in one direction, characterised in that the fibrils of P1 are flat and generally parallel with the main surfaces of the film with thicknesses generally around or lower than 1 μm , and width at least 5 times the thickness and further

5 characterised in that P1 is chosen to exhibit desirable barrier properties and further characterised by comprising, in longitudinal cross-section perpendicular to the main surfaces of the film, at least 4 die lines.

10 57. A film according to claim 56, characterised by a minor coextruded surface layer on at least one side of the alloy layer to enhance bonding properties and/or modify its frictional properties.

58. A film according to claim 56, characterised in that P1 consists of EVOH, a vinylidene chloride copolymer or polyamide.

15 59. An extruded film according to claim 56 which is uniaxially or biaxially oriented and is laminated to another oriented film, whereby the main directions of orientation cross each other.

20 60. A cellular expanded film made by extrusion in the presence of an expansion agent, characterised in that the film is made from an alloy of at least two polymers P1 and P2, which both are at least partly crystalline at temperatures under 100°C, the alloy comprising a dispersion of microscopically fine fibrils or fibril network of P1 surrounded by P2, whereby each fibril extends mainly in one direction and is flat with thicknesses generally around or lower than 1 μm , and width at least 5 times the thickness.

25 61. A film according to claim 60 which is uniaxially or biaxially oriented and is laminated to another film, whereby the main directions of orientation cross each other.

62. A film according to claim 60 in the form of rope, twine or woven-tape products.

63. A film according to claim 60 in the form of split fibre products.

30 64. A film according to any of claims 60 to 63 in which wherein P2 in its unoriented state at 20°C exhibits a coefficient of elasticity (E) which is at least 15% lower than E of P1.

65. A film according to any of claims 56 to 65 in which P2 is a copolymer of propylene or polyethylene, preferably a copolymer of ethylene and another alphaolefin, preferably LLDPE.

66. A film according to any of claims 56 to 66, in which, in the alloy, 5 the weight proportion of P1 is in the range 5 to 75 %.

67. Apparatus for extruding thermoplastic material comprising a die having an exit orifice through which the molten material flows and stretching means for stretching the material after it is extruded by at least two steps, in the first of which the material is stretched longitudinally by first stretching 10 means whilst at a high temperature, and in the second of which the material is stretched longitudinally by second stretching means at a lower temperature, comprising also means for cooling the extruded material between the two stretching means, said cooling means comprising a frictional device arranged for contact with the extruded material, 15 characterised by further comprising stretching means downstream from said second stretching means, and additional cooling means between said second stretching means and said further stretching means.

68. Apparatus according to claim 67 in which the said frictional device is provided with holes or is made of microporous metal for inwards or 20 outwards passage of air whereby over or under pressure of air is provided to control the friction between the device and the material.

69. Apparatus according to claim 67 or 68 comprising a shock cooling part upstream of the frictional device past which the extruded flow passes and which is cooled by a flow of cooling medium through its interior.

70. Apparatus according to claim 69 which further comprises 25 heating means between the shock cooling means and the frictional device, for controlled heating of the material.

71. Apparatus according to claim 70 in which the heating means comprises a pair of fixed heating blocks arranged on opposite sides of the 30 extruded material.

72. Apparatus according to any of claims 67 to 71 in which the die has a grid chamber upstream from the exit orifice comprising one or more grids through which the extrudate passes, the grid or grids being located at a position in the chamber where the gap is wider than said exit orifice gap, the grid chamber further comprising a gap reduction portion between the grid or grids and the exit orifice wherein the gap is reduced at least part way to the gap of the exit orifice.

73. Apparatus for extruding thermoplastic material comprising a die having an exit orifice through which the molten material flows and stretching means for stretching the material after it is extruded by at least two steps, in the first of which the material is stretched longitudinally by first stretching means whilst at a high temperature, and in the second of which the material is stretched longitudinally by second stretching means at a lower temperature, comprising also means for cooling the extruded material between the two stretching means, said cooling means comprising a frictional device arranged for contact with the extruded material, characterised in that there is provided a grid chamber upstream from the exit orifice comprising one or more grids through which the extrudate passes, the grid or grids being located at a position in the chamber where the gap is wider than said exit orifice gap, the grid chamber further comprising a gap reduction portion between the grid or grids and the die exit wherein the gap is reduced at least part way to the gap of the exit orifice.

74. Apparatus according to claim 72 or 73 in which each such grid has walls extending several mm in the direction of the flow.

75. Apparatus according to any of claims 73 to 74 in which the major walls in each such grid are substantially planar and are slanted so that each forms an angle between about 10 to 70° to the major surface of the extrudate flow entering the grid.

76. Apparatus according to claim 75 in which said angle and the wall thickness and distances between the walls are such that, in a

longitudinal section of the die perpendicular to the main surfaces of the extrudate flow as this enters the grid, there are at least four such walls.

77. Apparatus according to claim 75 or 76 which comprises at least two such grids which are slanted in opposite directions to one another.

5 78. Apparatus according to any of claims 67 to 77 comprising means for coextruding a surface layer at least on one side of the extrudate.

79. Apparatus according to any of claims 67 to 78 comprising means for transverse stretching of the extruded film downstream of the second stretching means.

10 80. Apparatus according to claim 79 in which upstream of the transverse stretching means there is a longitudinal pleating device, preferably comprising a pair of rubber belts between which the extruded material passes.

15 81. Apparatus according to any of claims 79 to 80, in which the transverse stretching means comprises a tenterframe including an oven.

82. Apparatus according to claim 81, in which said oven comprises fixed heated blocks arranged on opposite sides of the material, provided with heating means, preferably electrical heating means.

20 83. Apparatus according to claim 82, which further comprises a cooling block on at least one side of the material, downstream of the heating blocks, said cooling block being provided with a channel for passage of cooling air, preferably in which said channel is in fluid communication with the surface of the block facing the extruded material, by virtue of forming the cooling block from microporous metal.

25 84. Apparatus according to claim 82 or 83, in which said heating blocks are formed of microporous metal in fluid contact with channels for passage of heated air, whereby heated air exits the blocks from the surfaces facing the material passing therebetween, to lubricate passage of the material therebetween.

30 85. Apparatus according to any of claims 67 to 72, in which the further stretching means is a longitudinal stretching means and the

apparatus preferably comprises pleating means for imposing transverse pleats in the material prior to said longitudinal stretching.

86. Apparatus according to claim 85, including a laminating station, in which a second sheet material is laminated to the extrudate, said 5 laminating station preferably being upstream from the longitudinal cold stretching means.

87. Apparatus according to claim 86, in which the extrusion die is a circular die for extruding a tube of material, and which further comprises helical cutting means downstream of the said second stretching station, and 10 upstream of the laminating station, in which the tube of material is helically cut and two plies of the extruded material are laminated to one another with their main directions of orientation arranged at an angle to one another.

88. Apparatus according to any of claims 67 to 86 in which the extrusion die is a flat die.

89. Apparatus for extruding thermoplastic material comprising a die 15 having an exit orifice through which the molten material flows and stretching means for stretching the material after it is extruded, characterised in that the die has a grid chamber upstream from the exit orifice comprising one or more grids through which the extrudate passes, the grid or grids being 20 located at a position in the chamber where the gap is wider than said exit orifice gap, the grid chamber further comprising a gap reduction portion between the grid or grids and the exit orifice wherein the gap is reduced at least part way to the gap of the exit orifice and the or each grid comprises at 25 least 4 (in the longitudinal sections perpendicular to the main surfaces of the flow) closely spaced lamellae having walls extending several mm in the direction of flow of molten material and, between the lamellae having apertures through which the molten material can flow.

90. Apparatus according to claim 89 in which the lamellae in each such grid are slanted so that each forms an angle between about 10 to 70° 30 to the major surface of the extrudate flow entering the grid.

91. Apparatus according to claim 90 in which the lamellae are substantially planar and preferably are substantially parallel to the flow as it enter the grid.

92. Apparatus according to claim 90 or 91 which comprises at least 5 two such grids which are slanted in opposite directions to one another.

93. Apparatus according to claim of claims 89 to 92 comprising means for coextruding a surface layer at least on one side of the extrudate.

94. Apparatus according to any of claims 89 to 93 in which the extrusion die is a circular die for extruding a tube of material.

95. Apparatus according to any of claims 89 to 93 in which the extrusion die is a flat die.

96. A film according to claim 49a, 56 or 60 in which the widths of the fibrils are at least 10 times the thickness.

97. A film according to claim 49 which , in longitudinal cross-15 section perpendicular to the main surface of the film, comprises at least 4 die lines.

STATEMENT UNDER ARTICLE 19 (1)

The claims are amended under Article 19 PCT.

Claim 1 has been amended to clarify the wording. Claim 32 is amended to recite additional features of the grid. Claim 32 is also amended to refer to the step of stretching of the film after extrusion from the die.

The amendments in claim 32 to define the grid further serve to clarify the distinction from JP-A-57-110422. Claims 32 is distinguished also from US-A-2622989 by virtue of the further definition of the grid, specifically the requirement that this be formed of lamellae.

Original claim 33 has been deleted. Original claim 34 is split into two claims (33 and 34) with amendments consequential upon the reference to lamellae in claim 32.

Original claim 35 is amended as a consequence of the amendment to claim 32 and refers to the feature described on page 11 line 16.

A minor clarifying amendment is made in claim 43.

Claim 49 has a clarifying amendment in part b and has been limited to the products of claim 54 or 55. This claim is distinguished from EP-A-0337719, since it relates to oriented films. The prior art does not disclose orienting the films after extrusion. The claim is distinguished from US 2,262,989 by the same feature, the requirement for the film to be oriented. It appears to be intended that the film produced in US 2262989 is not oriented. The requirement that the film be oriented distinguishes the product also from that produced in JP-A-57-110422. None of the references mentions the product forms to which this claim is now limited.

New claim 97 has been added which is, a claim dependent on claim 49, which specifies that the film comprise at least 4 die lines. The die lines are discussed on page 29 line 22 to page 30 line 2. The die lines are the result of passing the extrudate through grids as defined in claim 32. The die lines are intentionally avoided in JP-A-57-110422, so this claim provides a further distinction from that prior art. Die lines would also not be the inevitable result of the processes described in EP-A-0337719 and US 2,262,989.

Claim 56 is amended, to change the definitions of the components P1 and P2, and is based on description page 22, product B the original formulation of the claim inappropriately referred to the coefficient of elasticity of the components. This

claim has been further amended to delete reference to a preferred feature. It has furthermore been limited to specify the feature that the film, in cross-section, comprises at least 4 die lines. The claim is distinguished from each of the prior art references mentioned above by virtue of this requirement for die lines, for reasons explained in relation to claim 97.

Claim 60 is not amended. It is distinguished from the prior art mentioned above by virtue of the requirement for extrusion in the presence of an expansion agent. Expansion agents are not mentioned in the prior art listed above.

New apparatus claim 89 has been added which corresponds substantially to claim 32. The claim is patentable over the prior art relevant to original claim 32 for reasons explained above in relation to claim 32 as amended. Dependent claims 90-95 correspond substantially to the preferred features of the apparatus, based on dependent apparatus claims 75-78, 87 (at first part) and 88.

New claim 96, a dependent product claim, defines preferred features of the fibrils in the film.